PROCESS FOR PRODUCING OLEFIN POLYMERS

Background of the Invention

5   **Field of the Invention**

The present invention relates to olefin polymerization. In particular, the present invention concerns a process for polymerization of propylene in a polymerization reactor arrangement to produce at least two olefin polymers having essentially the same level of Melt Flow Rate

10   but different isotacticity.

Description of Related Art

Polypropylenes are conventionally produced in the presence of a high-yield catalyst system

15   comprising a catalyst component, a cocatalyst component and a compound acting as an external electron donor. The catalyst component is typically a supported Ziegler-Natta type catalyst containing magnesium, titanium and a halogen as essential components. Porous, inorganic or organic particulate carrier materials, such as silica or $MgCl_2$ supports, are used as support materials. The external electron donors present a means for controlling the isotacticity

20   of the polymer.

For different applications, polymers with very different properties are required. The main characteristics of these polymers are their isotacticity and Melt Flow Rate, in the following also abbreviated "MFR". These features can be controlled by varying the process conditions

25   and by using different catalyst systems. One important tool for adapting the properties of the polymers to varying product requirements is adjustment of hydrogen feed during polymerization. By varying of the feed of hydrogen, the molecular weight or MFR of the polymer can be controlled. The stiffness of the polymer is also an important product property, which should be adjusted depending on the end use of the polymer. Stiffness is greatly

30   dependent on the isotacticity of the polymer and, thus, isotacticity has to be set at the desired, predetermined level. This is typically done by proper selection of the external donors used in the polymerization process.

It is commonly known that different external donors lead to polymers with different isotacticities and to polymers having different mechanical properties. One of the important mechanical properties is the flexural modulus. It is also known that the isotacticity can be affected to some extent by changing the concentration of the donor. This change in donor

5   concentration influences the concentration of xylene-solubles of the polymer, which is usually not desired.

There are problems related to the known processes, when transitions from one polymer grade to another grade have to be made. It is often necessary to produce different polymer grades

10   with the same process equipment. With prior art processes it is extremely difficult, and sometimes even impossible, to change over from one polymer grade to another by changing one polymer characteristic without affecting the other polymer characteristics. According to the prior art, the change in one feature or one component of the process tends to cause changes in other features, too. This means in practice that if one component or feature in a

15   process for changing one property of the product is changed, then one or more other components of the process have to be changed, too, if the other properties of the polymer are to be kept unchanged. This is due to the fact that components of the process are so closely linked together that changes in one component mean changing the others, too. This is a great problem with present-day processes, where the transition of polymer grades having different

20   stiffness, but a predetermined MFR has to be made.

As is stated above, different isotacticity levels require the use of different external donors. However, hydrogen response of the catalyst system is dependent on the type of external donors used. "Hydrogen response" or "hydrogen sensitivity" stands for the sensitivity of the

25   molecular weight of the polymer to the hydrogen concentration. This means that if the external donor is changed, the hydrogen feed to the process needs to be recalculated and changed in order to meet the requirements for a specific level of melt flow rate. As far as the process is concerned, this causes extra work, time and costs.

30   Thus, in summary, according to known processes, when isotacticity and MFR of the polymer are modified, the donor needs to be changed and the hydrogen flow to the process needs to be altered. Even if only isotacticity is to be changed by changing the donor, also the hydrogen feed has to be changed due to the reasons explained above. Further, there is always a transitional period of time before the grade of the polymer product is changed from a first

polymer grade to another, since conventional processes stabilize slowly after a change of donor and hydrogen feed. The material produced during the switching-over period between two grades often has to be discarded because it does not fulfil the quality requirements neither of the first nor of the second polymer. This is both an economical and an environmental

5   disadvantage.

Until now, no method of transition from one polymer grade to another is known, wherein the hydrogen feed could be kept on the same level when an external donor is changed, in order to change the isotacticity, but to keep the MFR at the predetermined level. In practice, this

10   means that there are no processes that would allow for facile transition from one polymer grade to another having different isotacticity but essentially the same MFR.

Summary of the Invention

15   It is an aim of the present invention to eliminate the problems of the prior art and to provide a novel way of producing two or several grades of propylene polymers, including propylene homo- and copolymers, in one and the same polymerisation reactor system, while avoiding extended transitional periods between the change from one grade to another.

20   It is another object of the invention to provide a propylene polymerization process, where the control of isotacticity and MFR of the propylene polymer can be optimised during the production of propylene homopolymers and/or propylene copolymers comprising propylene random and heterophasic (block) copolymers.

25   Furthermore, a third object of the invention is to provide a process, where the sensitivity of the molecular weight of the polymer to the hydrogen concentration (i.e. hydrogen response or sensitivity of the catalyst) is increased. Such a process will provide for a novel means of controlling isotacticity of polypropylene polymers primarily only by using external donors.

30   These and other objects, together with the advantages thereof over known processes and products, are achieved by the present invention as hereinafter described and claimed.

The invention is based on the surprising finding that a specific, newly-developed kind of catalysts can be used for producing, at essentially the same polymerization conditions,

different polymers having the same, predetermined level of MFR but different isotacticity. These new catalysts will allow for the same hydrogen response while using different external donors.

5   Typically, the catalysts comprise particles of Group 3 to 10 transition metal compounds, or an actinide or lanthanide, in combination with compounds or complexes of Group 2 metals, produced by solidification of particles from emulsion systems to produce catalyst particles having a desired particle size. In particular, the catalyst particles are obtained by forming a liquid-liquid emulsion system, which contains a homogeneous solution of the at least one

10   catalyst component, said solution being dispersed in a solvent immiscible therewith and forming the dispersed phase of the liquid-liquid emulsion system, solidifying said dispersed droplets to form solid catalyst particles having a predetermined size range, and recovering said solid catalyst particles. The particles are "self-supporting" in the sense that they are not supported on any external carrier. An essential feature of the obtained catalyst is that the

15   active sites of the catalyst are evenly distributed thorough the whole particles contrary to the normally used commercial supported ZN catalysts, where the surface of an external support is treated with the catalyst, whereby the active sites of the catalyst are concentrated only on the surface of the support or carrier.

20   These novel kinds of catalysts and their preparation are described in WO Publications Nos. 03/000754 and 03/000757, the contents of which are herewith incorporated by means of reference.

       The above-described catalysts can, according to the present invention, now be used in a novel

25   transition process for producing at least two different propylene polymer grades, in which process the isotacticity of the polymer is changed while keeping the melt flow rate of the polymer at a predetermined level during a transition of production – in particular continuous production – from a first polymer grade to a second, said process being carried out in a polymerization arrangement comprising at least one polymerization reactor, where propylene

30   is polymerized, optionally with comonomers, under polymerization conditions in the presence of hydrogen as a molecular weight controlling agent and a Ziegler-Natta catalyst system, comprising a catalyst component and an external donor, wherein the external donor is changed, but the hydrogen feed is kept at a predetermined level, during a transition of production from the first polymer grade to the second.

In practice, the production of the first polymer grade is carried out in the presence of a catalyst system comprising a first external donor and the production of the second polymer grade is carried out in the presence of a catalyst system comprising a second (different) external donor. According to the invention, the change of the external donor comprises exchanging the first

5   external donor for the second external donor.

The above-described catalysts can also be used in a process for producing at least two olefin polymers having essentially the same level of MFR but different isotacticity, by polymerizing the olefin monomers in the presence of the catalytic system and by maintaining the hydrogen

10   feed at an essentially constant level during the production of at least the first and the second polymers.

Further, the catalysts provide for a process of controlling the isotacticity of polypropylene polymers by using external donors, in which process the isotacticity is adjusted by changing

15   the external donor without changing the hydrogen feed and still maintaining the melt flow rate essentially at the same level.

More specifically, the process according to the present invention is mainly characterized by what is stated in the characterizing part of claims 1, 7 and 25.

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The present invention provides important advantages. Thus, the present invention makes it faster and easier to control the overall process and especially to change from one polymer grade to another, while maintaining good control over the important properties, like MFR and isotacticity, of the polymer product. As pointed out above, with known processes, transfer

25   from producing one polymer grade having a predetermined MFR level to produce another polymer grade having the same MFR level but different isotacticity is often difficult. The hydrogen feed has to be changed and this operation will lead to a lowered production rate in order to decrease the off-spec material, the amount of which might be undesired high in conventional processes. This is, as stated above, due to the fact that a change from one

30   external donor to another leads to different hydrogen response and, thus, to necessary changes in hydrogen feed. This is, however, not necessary for the invention. On the contrary, it is an essential and important feature of the invention that the hydrogen feed can be maintained on essentially the same (predetermined or preset) level during the entire operation of the

polymerization process comprising the change of the external donor for producing two or more polymers having different isotacticity but the same MFR.

Another great advantage is that the transition from one grade of polymer to another is faster
5   and easier, because there is no need to alter the hydrogen feed to the process. In other words, the hydrogen feed curves do not have to be recalculated and reoptimized when the polymer grade is changed. There will be less rejected material that does not fulfil product requirements, because the target level of product properties is reached sooner. This is an advantage when thinking of both economical (lower operational cost) and environmental
10   aspects. In addition to this, a broader product window can be reached much easier.

A further problem associated with prior art processes is that the final stages of product development are time-consuming and expensive. New grades have to be tested at real process conditions using real process equipment. With the help of the present invention, product .
15   development will be faster. Product development involves testing new polymer grades and often involves switching from one donor to another. With prior art processes, this means changing the hydrogen flow. With help of the present invention, the time requirement for the switch is shorter, because there is no need to alter the hydrogen feed to the process.

20   In polymerization processes, control systems play an important role. For prior art processes, there are many variables to control. One of them is the hydrogen feed, which usually needs to be altered when polymer grades are changed. The more variables there are to control, the more difficult it is to reach the desired properties when the variables are changed. In other words, there is more off grade produced. Also transition times are longer with additional
25   parameters to be changed in product change. The present invention allows for easier control of the overall process because there is one variable less to control and may lead to lower investment cost.

With the aid of the present invention, it is possible to produce a wider product range or
30   product window with the same process. By using the process of the present invention, it is possible to produce propylene polymers having a Melt Flow Rate (MFR_2) 0.01 to 1500 g/10, min or even higher, (measured by ISO Standard 1133, at 230 °C, 2.16 kg load) and isotacticity in the range, which is normally obtainable by donors used in the propylene polymerization processes.

Hydrogen feed is maintained at essentially constant level, when making polymer with same MFR, but by using different external donor, i.e. the feed is independent on the donor. For the purpose of this invention, "essentially constant level" means that the volume of the hydrogen

5   feed will change at the most 5 % from the starting volume. The level of the hydrogen feed is chosen to produce polymers with desired MFR. Further details and advantages of the invention will become apparent from the following detailed description comprising a number of working examples.

10   **Brief Description of the Drawings**

Figure 1 depicts graphically the results of polymerization experiments disclosed in Examples 1, 2 and 5 according to the invention. The x-axis represents hydrogen flow and the y-axis MFR_2 of the polymer.

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Figure 2 depicts graphically the result of polymerization experiments of Comparative Examples 1, 2 and 3. The x-axis represents hydrogen flow and the y-axis MFR_2 of the polymer.

20   **Detailed Description of the Invention**

Overall Process

Generally, the present invention comprises the steps of producing

25   –   a first polymer or polymer grade with a predetermined MFR and a first degree of isotacticity in the presence of said catalytic system using a first external donor; and

–   a second polymer or polymer grade with essentially the same predetermined MFR and a second degree of isotacticity in the presence of said catalytic system using a second external donor. The second donor can be the same as or, preferably,

30   different from the first donor.

The process steps can be performed in optional order and in a sequence one after another. The process can be a one stage or a multistage polymerization process carried out in liquid/slurry phase, gas phase or vapour phase in a sequence of polymerization steps comprising one or

more liquid/slurry polymerization and/or one or more gas phase polymerization steps or combinations thereof in any order. The polymerization in each stage can be carried out in one or more reactors such as slurry/bulk polymerization reactors and or gas phase reactors. In one embodiment the polymerization is carried out in at least one bulk reactor, preferably loop

5   reactor, followed by at least one gas phase reactor.

Polymers

The polymers produced with the process of the invention are propylene polymers comprising

10   propylene homopolymers, propylene copolymers comprising propylene random and heterophasic (block) copolymers or combinations thereof.

The comonomers used are selected from the group of C_{2-18} olefins, preferably C_{2-10} olefins, such as ethylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 1-

15   nonene and 1-decene as well as mixtures thereof and dienes, such as 1,5-hexadiene and 1,9-decadiene. Most commonly used comonomers are ethylene and 1-butene, especially ethylene.

By using the process of the present invention, it is possible to produce propylene polymers having a Melt Flow Rate (MFR_2) of 0.01 to 1500 g/10 min or even higher, preferably 0.1 to

20   500 g/10 min, in particular 10 to 300 g/min, (measured by ISO Standard 1133, at 230 °C, 2.16 kg load). As examples of some particularly preferred ranges the following can be mentioned (measured as explained above): 20 to 200 g/min, 20 to 100 g/min, 50 to 150 g/min and 100 to 300 g/min, depending on the final application. FTIR Isotacticity (measured by FTIR , Fourier transform infrared spectra, ^{13}C NMR calibrated) depends, as is well known, on the MFR of

25   the polymer. As examples of the higher FTIR isotacticity-values of homoPP produced by the process of the invention are e.g. as follows: at MFR_2 of 0,3 g/10 min about 91 %, at MFR_2 of 20 g/10 min about 99 % and at MFR_2 of 300 g/10 min about 103 %. Lower isotacticity (= higher XS), at a certain MFR-level, can be achieved by decreasing donor concentration or by using a weaker external donor in the polymerisation. However, changing the donor

30   concentration will also change the hydrogen sensitivity of the catalyst system, which causes a need to alter also hydrogen feed. In this case, two changes in the process are needed. If the lower isotacticity is achieved by changing the external donor, only one change in the process is needed, because hydrogen sensitivity of the catalyst system does not change even if the external donor is changed.

According to the present invention, a smooth transition from one grade of polymer to another grade is now possible;.polymers having the same MFR level, but different isotacticities can be obtained without changing the hydrogen feed from the polymerization of the first polymer to the following polymer(s). The process provides continuous polymerization of propylene monomers along with any possible comonomers using a catalyst having essentially the same hydrogen response independent on the used external donors. This means that the catalyst used exhibits the same hydrogen response and, thus, the MFR of the polymers will remain essentially unchanged, although different isotacticities are desired and obtained. As a result, the switch or transition from one grade to another will be facile and rapid, resulting in no or only minor losses in production output during the transition period.

According to an embodiment of the invention a first polymer is produced, having a predetermined MFR and a lower isotacticity and a second polymer is produced having the same MFR but a higher isotacticity. This sequence can be followed by a third and a fourth and several further polymers having even higher or again lower isotacticities than any of the previous polymers.

According to another embodiment, the isotacticity of the second polymer may be lower than that of the first polymer. This sequence can be followed by a third and a fourth and several further polymers having even lower or again higher isotacticities than any of the previous polymer.

Reactors

The polymerization reactor arrangement comprises one or more liquid/slurry/ reactor(s), preferably bulk reactor(s), or one or more gas phase reactor(s), or combinations of any of these in any order. The reactors can be arranged in series. The polymerization may be carried out in several stages each comprising polymerizations in one or more reactors.

The first stage polymerization can be carried out in one or more bulk reactor(s), preferably loop reactor, or in one or more gas phase reactor(s). Typically, the reactors are connected in series. The second stage polymerization is preferably carried out in one or more gas phase reactor(s). The second stage polymerization is typically carried out essentially immediately

after the first stage polymerization, i.e., the polymerization product recovered from the first polymerization stage is conducted to the first gas phase reactor of the second polymerization stage. The gas phase reactors in the second polymerization stage are preferably connected in series.

The polymerization temperature in the actual polymerizations is usually in the range of 60 to 110 °C, preferably between 70 and 100 °C, more preferably between 70 and 90°C and most preferably between 70 and 85°C. The pressure in the slurry/bulk reactors, preferably in loop reactors is typically between 20 to 70 bar, preferably between 30 to 60 bar. The pressure in the gas phase reactors is typically between 10 to 40 bar, preferably 15 to 30 bar.

According to one embodiment, the heterophasic propylene copolymer is produced in a reactor system comprising at least one liquid (bulk) reaction stage including at least one liquid (bulk) reactor, preferably at least one loop reactor, and at least one gas phase reaction stage including at least one gas phase reactor. The reactors are connected in series, the bulk reactors being preferably arranged before the gas phase reactors. A separation stage can be employed between the reaction stages or reactors to prevent the carryover of reactants from the one polymerization stage or reactor into another.

According to this embodiment the polymer matrix is produced in the first reaction stage, which is preferably a bulk reaction stage, and the heterophasic rubbery part is produced in the second reaction stage, which preferably is a gas phase reaction stage.

In addition to the actual polymerization reactors used, the polymerization reaction system can also include a number of additional reactors, such as prereactors. The prereactors include any reactor for preactivating and/or prepolymerizing the catalyst e.g. with propylene and/or other α-olefin(s) and/or ethylene, if necessary. In addition the polymerization system can contain some post-reactors for further modifying the produced polymer. All reactors in the reactor system are preferably arranged in series.

According to the invention, it is not necessary to change the hydrogen feed to keep the MFR of the polymer at the predetermined level, although adjustment of the isotacticity is aimed at, e.g. by changing the external donor used to another type of donor. I.e. the hydrogen feed can

be maintained at an essentially constant level in the polymer transition process. For the purpose of this invention, "essentially constant hydrogen feed level" means that during the transition from a first polymer to another, the predetermined MFR can be maintained and the desired isotacticity of the second or further polymer can be reached without altering the

5   volume of the hydrogen feed. Keeping the hydrogen feed essentially at the same level, means that it will be changed no more than 5 %, preferably no more than 3 %, most preferably no more than 2 %, from the starting volume. The level of the hydrogen feed is chosen so as to enable production of a first polymer with a desired MFR and isotacticity and a second or following polymers with the same, predetermined MFR but different isotacticity.

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The catalyst

The catalyst system comprises a catalyst component, optionally a procatalyst component and a cocatalyst component, and an external donor.

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The external donors used in the present invention are strongly coordinating donors, which form relatively strong complexes with catalyst surface, mainly with $MgCl_2$ surface in the presence of aluminium alkyl and $TiCl_4$. The donor components are characterised by a strong complexation affinity towards catalyst surface and a sterically large and protective

20   hydrocarbon. Strong coordination with $MgCl_2$ requires oxygen-oxygen distance of 2.5 to 2.9 Å [Albizzati et al., Macromol. Symp. 89 (1995) 73-89].

Typically this kind of donors are silane-based donors having the structure of the general formula I

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$$R'''_nSi(OMe)_{4-n} \hspace{4cm} (I)$$

wherein R''' is a branched aliphatic or cyclic or aromatic group, Me stands for methyl and n is 1 or 2, preferably 2. [Härkönen et al., Macromol. Chem. 192 (1991) 2857-2863].

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In particular, the external donor is selected from the group consisting of dicyclopentyl dimethoxysilane (donor D), cyclohexylmethyl dimethoxy silane (donor C), diisopropyl dimethoxysilane, methylcyclohexyldimethoxy silane, di-isobutyl dimethoxysilane, and di-t-butyl dimethoxysilane. Dicyclopentyl dimethoxysilane (donor D) and cyclo hexyl methyl

dimethoxy silane (donor C) are particularly preferred. From these donors D and C, donor D has, according to common knowledge, a stronger coordinating effect than donor C.

The present catalyst system comprises catalyst particles, which, according to the present
5   invention, are of a specific constitution in the sense that they are not supported on any external carrier contrary to the normally used commercial supported ZN catalysts. The active sites of the catalyst are evenly distributed throughout the whole particles and the particles are "self-supporting". Catalysts of this kind can be prepared as described in WO Publications Nos. 03/000754 and 03/000757.
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In addition to the above-discussed features, the catalyst particles prepared according to the cited documents have excellent morphology and good, uniform particle size distribution and due to the replica effect the polymer particles produced by using these catalysts have very good morphology properties, too.
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The olefin polymerization catalyst component used in the present invention, comprises a compound of a transition metal of Group 3 to 10 of the Periodic Table (IUPAC), or of an actinide or lanthanide, and is prepared according to a method comprising

(a)   forming a liquid/liquid two phase emulsion system, which contains a homogeneous
20   solution of at least one catalyst component, said solution being dispersed in a liquid medium and forming the dispersed phase of the liquid/liquid emulsion system,

(b)   solidifying said dispersed droplets to form solid catalyst particles having a predetermined size range,

(c)   recovering the obtained solidified catalyst particles.
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As regards the "predetermined size range", it should be noted that the catalyst particles usually have an average size in the range of 5 to 200 µm, preferably 10 to 100, more preferably 20 to 50 µm.

30   The catalyst component can include, in addition to said transition metal compound, also any additional cocatalyst(s) (e.g. additional transition metal compounds) and/or activators and/or poison scavengers) and/or any reaction product(s) of a transition compound(s) and a cocatalyst(s). Thus the catalyst may be formed *in situ* from the catalyst components in said solution in a manner disclosed in said references.
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A preferred process for producing an ZN propylene polymerization catalyst component in the form of particles having a predetermined size range comprises preparing a solution of a complex of a Group 2 metal and an electron donor by reacting a compound of said metal with said electron donor or a precursor thereof in an organic liquid reaction medium; reacting said

5 complex, in solution, with a compound of a transition metal to produce an emulsion, the dispersed phase of which contains more than 50 mol-% of the Group 2 metal in said complex; maintaining the particles of said dispersed phase within the average size range 5 to 200 μm by agitation preferably in the presence of an emulsion stabilizer and solidifying said particles; recovering and optionally washing said particles to obtain said catalyst component.

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For said ZN catalyst particles, the compound of a transition metal is preferably a compound of a Group 4 metal. The Group 4 metal is preferably titanium, and its compound to be reacted with the complex of a Group 2 is preferably a halide. In a still further embodiment of the invention a compound of a transition metal can also be selected from Group 5 metals, Group

15 6 metals, Cu, Fe, Co, Ni and/or Pd. The complex of the Group 2 metal is preferably a magnesium complex.

The liquid medium used in the formation of the liquid/liquid two phase emulsion system is a medium being immiscible to the solution of at least one catalyst component at least to the

20 extent that an emulsion can be formed.

In a preferred embodiment the process for producing catalysts used in the invention comprises: preparing a solution of magnesium complex by reacting an alkoxy magnesium compound and an electron donor or precursor thereof in a C_6-C_{10} aromatic liquid reaction

25 medium; reacting said magnesium complex with a compound of at least one four-valent Group 4 metal at a temperature greater than 10 °C and less than 60 °C, to produce an emulsion of a denser, $TiCl_4$/toluene-insoluble, oil-dispersed phase having, Group 4 metal/Mg mol ratio 0.1 to 10 in an oil disperse phase having Group 4 metal/Mg mol ratio 10 to 100; maintaining the droplets of said dispersed phase within the size range 5 to 200 μm by

30 agitation in the presence of an emulsion stabilizer while heating the emulsion to solidify said droplets and adding turbulence minimizing agent into the reaction mixture before solidifying said droplets of the dispersed phase, said turbulence minimizing agent being inert and soluble in the reaction mixture under the reaction conditions; and recovering the obtained olefin

polymerization catalyst component. The recovering step comprises removal of the solvent from the mixture, which can be done e.g. by drying or by other means.

In the above, the term "oil-dispersed phase" means that the phase containing the catalyst
5   component(s) is an oil-like liquid.

The turbulence minimizing agent (TMA) or mixtures thereof preferably comprises polymers having linear aliphatic carbon backbone chains, which might be branched with short side chains only in order to serve for uniform flow conditions when stirring. Said
10 TMA is in particular preferably selected from α-olefin polymers having a high molecular weight of MW about $1 - 40 \times 10^6$, or mixtures thereof. Especially preferred are polymers of α-olefin monomers with 6 to 20 carbon atoms, and more preferably polyoctene, polynonene, polydecene, polyundecene or polydodecene or mixtures thereof, having the molecular weight and general backbone structure as defined before, and most preferable
15 TMA is polydecene.

As electron donor compound to be reacted with the Group 2 metal compound is preferably an mono- or diester of an aromatic carboxylic acid or diacid, the latter being able to form a chelate-like structured complex. Said aromatic carboxylic acid ester or diester can be
20 formed in situ by reaction of an aromatic carboxylic acid chloride or diacid dichloride with a C_2-C_{16} alkanol and/or diol, and is preferable dioctyl phthalate, such as di-ethyl-hexyl phthalate. The liquid reaction medium preferably comprises toluene.

The reaction for the preparation of the Group 2 metal complex is generally carried out at a
25 temperature of 20° to 80°C, and in case that the Group 2 metal is magnesium, the preparation of the magnesium complex is carried out at a temperature of 50° to 70°C.

The emulsion stabiliser is typically a surfactant, of which the preferred class is that based on acrylic polymers.
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The alkoxy magnesium compound group is preferably selected from the group consisting of magnesium dialkoxides, complexes of a magnesium dihalide and an alcohol, and complexes of a magnesium dihalide and a magnesium dialkoxide. It may be a reaction product of an

alcohol and a magnesium compound selected from the group consisting of dialkyl
magnesiums, alkyl magnesium alkoxides, alkyl magnesium halides and magnesium dihalides.
It can further be selected from the group consisting of dialkyloxy magnesiums, diaryloxy
magnesiums, alkyloxy magnesium halides, aryloxy magnesium halides, alkyl magnesium

5   alkoxides, aryl magnesium alkoxides and alkyl magnesium aryloxides.

The magnesium dialkoxide may be the reaction product of a magnesium dihalide such as
magnesium dichloride or a dialkyl magnesium of the formula R_2Mg, wherein each one of the
two R's is a similar or different C_1-C_{20} alkyl, preferably a similar or different C_4-C_{10} alkyl.

10   Typical magnesium alkyls are ethylbutyl magnesium, dibutyl magnesium, dipropyl
magnesium, propylbutyl magnesium, dipentyl magnesium, butylpentylmagnesium, butyloctyl
magnesium and dioctyl magnesium. Most preferably, one R of the formula R_2Mg is a butyl
group and the other R is an octyl group, i.e. the dialkyl magnesium compound is butyl octyl
magnesium.

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Typical alkyl-alkoxy magnesium compounds RMgOR, when used, are ethyl magnesium
butoxide, butyl magnesium pentoxide, octyl magnesium butoxide and octyl magnesium
octoxide.

20   Dialkyl magnesium, alkyl magnesium alkoxide or magnesium dihalide can react with a
polyhydric alcohol $R'(OH)_m$, or a mixture thereof with a monohydric alcohol R'OH.

Typical C_2 to C_6 polyhydric alcohols may be straight-chain or branched and include ethylene
glycol, propylene glycol, trimethylene glycol, 1,2-butylene glycol, 1,3-butylene glycol, 1,4-

25   butylene glycol, 2,3-butylene glycol, 1,5-pentanediol, 1,6-hexanediol, 1,8-octanediol, pinacol,
diethylene glycol, triethylene glycol, and triols such as glycerol, methylol propane and
pentareythritol. The polyhydric alcohol can be selected on the basis of the activity and
morphology desired to the catalyst component.

30   The aromatic reaction medium may also contain a monohydric alcohol, which may be
straight- or branched-chain. Typical C_1 -C_{20} monohydric alcohols are methanol, ethanol, n-
propanol, iso-propanol, n-butanol, iso-butanol, sec.butanol, tert.butanol, n-amyl alcohol, iso-
amyl alcohol, sec.amyl alcohol, tert.amyl alcohol, diethyl carbinol, akt. amyl alcohol, sec.
isoamyl alcohol, tert.butyl carbinol. Typical C_6-C_{10} monohydric alcohols are hexanol, 2-ethyl-

lbutanol, 4-methyl-2-pentanol, 1-heptanol, 2-heptanol, 4-heptanol, 2,4-dimethyl-3pentanol, 1-octanol, 2-octanol, 2-ethyl-1-hexanol, 1-nonanol, 5-nonanol, diisobutyl carbinol, 1-decanol and 2,7-dimethyl-2-octanol. Typical >C_{10} monohydric alcohols are n-1-undecanol, n-1-dodecanol, n-1-tridecanol, n-1-tetradecanol, n-1-pentadecanol,1-hexadecanol, n-1-

5   heptadecanol and n- 1 octadecanol. The monohydric alcohols may be unsaturated, as long as they do not act as catalyst poisons.

Preferable monohydric alcohols are those of formula R'OH in which R' is a C_2-C_{16} alkyl group, most preferably a C_4-C_{12} alkyl group, particularly 2-ethyl-1-hexanol.

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Preferably, essentially all of the aromatic carboxylic acid ester is a reaction product of a carboxylic acid halide, preferably a dicarboxylic acid dihalide, more preferably an unsaturated α,ß-dicarboxylic acid dihalide, most preferably phthalic acid dichloride, with the monohydric alcohol.

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The finally obtained catalyst component is desirably in the form of particles having an average size range of 5 to 200 µm, preferably 10 to 100, more preferably 20 to 50 µm and the active sites of the catalyst are evenly distributed in the whole catalyst particle.

20   The catalyst used in the present invention comprises a catalyst component prepared as aforesaid, in association with commonly used cocatalysts, preferably alkyl aluminium compounds, more preferably aluminium trialkyls or aluminium alkyl halids, most preferably trialkyl aluminium, such as triethyl aluminium and external donors, and is used for the polymerization of propylene optionally with other monomers, such as C_2 to C_{10}-olefins.

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Summarising what has been stated above, one particularly preferred embodiment of the invention comprises a process for controlling isotacticity of polypropylene polymers by using external donors, comprising

      —   feeding propylene together with optional comonomers along with hydrogen as a
30   molecular weight controlling agent and a Ziegler-Natta catalyst system, including a
            catalyst component having as essential components Ti, Mg and Cl cocatalyst, and an
            external donor, into a polymerization reactor arrangement formed by at least one
            polymerization reactor and

     —   polymerizing propylene together with the optional monomers under polymerization conditions in order to obtain a polymer product having a predetermined melt flow rate and isotacticity,

wherein isotacticity is adjusted by changing the external donor without changing the hydrogen feed and still maintaining the melt flow rate essentially at the same level.

The following non-limiting examples illustrate the invention in more detail:

Examples

In the examples, the following measurement methods were used:

MFR (Melt flow rate)   : ISO 1133, 230 °C, 2,16 kg load

XS (Xylene solubles)   : xylene soluble part at 25 °C , ISO 6427

T_m (Melting temperature): Differential scanning calorimetry, DSC, Mettler TA 820

FTIR isotacticity   : Fourier tranformed infrared spectra, ^{13}C NMR calibration

FLEXULAR MODULUS   : ISO 178

Example 1a

All raw materials were essentially free from water and air and all material additions to the reactor and the different steps were done under inert conditions in nitrogen atmosphere. The water content in propylene was less than 5 ppm.

The polymerization was done in a 5 litre reactor, which was heated, vacuumed and purged with nitrogen before taken into use. 348 μl TEA (triethyl aluminium from Witco used as received) as cocatalyst, 51 μl donor C (cyclo hexyl methyl dimethoxy silane from Wacker, dried with molecular sieves) and 30 ml pentane (dried with molecular sieves and purged with nitrogen) were mixed and allowed to react for 5 minutes. Half of the mixture was added to the reactor and the other half was mixed with 17,2 mg highly active and stereospecific Ziegler Natta catalyst (ZN catalyst). The ZN catalyst was prepared according to WO03/000754, example 7 and had Ti content 2.84 w-%. After about 10 minutes the ZN catalyst/TEA/donor C/pentane mixture was added to the reactor. The Al/Ti molar ratio was 250 and the Al/Do (Al/external donor) molar ratio was 10.

300 mmol hydrogen and 1400 g propylene were added to the reactor. The temperature was increased from room temperature to 70 °C during 20 minutes. The reaction was stopped, after 60 minutes at 70 °C, by flashing out unreacted propylene. Finally, the polymer powder was

5   taken out from the reactor and analysed and tested. The MFR of the product was 17. The details and results are shown in Table 1.

Example 1b

10   This example was carried out in accordance with Example 1a, with the exception that donor D (dicyclo pentyl dimethoxy silane) was used as external donor. The MFR of the product was 20, which is practically the same as that obtained with donor C in Example 1a. The details and results are shown in Table 1.

15

Example 2a

This example was carried out in accordance with Example 1a, with the exception that the amount of hydrogen was 1500 mmol. The MFR was 280 g/10min. The details and results are

20   shown in Table 1.

Example 2b

This example was carried out in accordance with Example 2a, with the exception that donor

25   D was used as external donor. The MFR was 290, which is practically the same as with donor C in Example 2a. The details and results are shown in Table 1.

Example 3a

30   This example was carried out in accordance with example 1a, with the exception that Al/Do ratio was 50. The MFR was 31. The details and results are shown in Table 1.

Example 3b

This example was carried out in accordance with Example 3a, with the exception that donor D was used as external donor. The MFR was 30, which practically is the same as

5   obtained with donor C in Example 3a. The details and results are shown in Table 1.

Example 4a

This example was carried out in accordance with Example 1a, with the exception that the

10   temperature in polymerization was 80 °C. The MFR was 24. The details and results are shown in Table 1.

Example 4b

15 · This example was carried out in accordance with Example 4a, with the exception that donor D was used as external donor. The MFR was 23, which is practically the same as in Example 4a, where donor C was used. The details and results are shown in Table 1.

Example 5a

20

This example was carried out in accordance with Example1a, with the exception that the amount of hydrogen was 6 mmol. The MFR was 0.32 g/10min. The details and results are shown in Table 1.

25   **Example 5b**

This example was carried out in accordance with Example 5a, with the exception that donor D was used as external donor, hydrogen amount was 7 mmol and polymerization temperature was 75 °C. The MFR was 0.32, which is practically the same as in example

30   5a, where donor C was used. The details and results are shown in Table 1.

Table 1. Results of Examples 1a to 5b

		Ex 1a	Ex 1b	Ex 2a	Ex 2b	Ex 3a	Ex 3b	Ex 4a	Ex 4b	Ex 5a	Ex 5b
Temperature	°C	70	70	70	70	70	70	80	80	70	75
Hydrogen	mmol	300	300	1500	1500	300	300	300	300	6	7
Donor	type	C	D	C	D	C	D	C	D	C	D
Al/Do	mol/mol	10	10	10	10	50	50	10	10	10	10
Activity	kg PP/ g cat h	37,2	28,2	36,4	30,7	46,5	33,8	30,5	27,9	13,4	10,2
MFR	g/10 min	17	20	280	290	31	30	24	23	0,32	0,32
XS	w-%	1,6	1,2	2,4	1,6	3,1	2,3	1,5	1,3	2,3	2
FTIR Isotacticity	%	97,6	98,7	101,2	103	95,9	97,3	98,8	99,4	91,2	90,9
Tm	°C	162	162,8	161,4	160,9	160,8	161,4	162,8	163,3	160,7	164,9
Flexural modulus	Mpa	1530	1660	1820	1800	1480	1550	1540	1640	1620	

5   **Comparative Example 1a**

This example was carried out in accordance with Example 1a, with the exception that the hydrogen amount was 13 mmol, polymerization temperature 80 °C, donor D was used as external donor and a different catalyst was used. The catalyst used in this test was a

10   transesterified $MgCl_2$ supported Ziegler Natta catalyst with Ti-content of 2,1 % for producing high stiffness polypropylene products. The catalyst was prepared in accordance with Finnish patent No. 88047. The MFR of the polymer was 0,36. The details and results are shown in Table 2.

15   **Comparative Example 1b**

This example was carried out in accordance with Comparative Example 1a, with the exception that donor C was used as external donor. The MFR was 0,70, which is 100 % higher than MFR in comparative example 1a, where donor D was used. The details and

20   results are shown in Table 2.

Comparative Example 2a

This example was carried out in accordance with Comparative Example 1a, with the

25   exception that the hydrogen amount was 200 mmol and polymerization time was 30 minutes. The MFR was 5.3. The details and results are shown in Table 2.

Comparative Example 2b

This example was carried out in accordance with Comparative Example 2a, with the
exception that donor C was used as external donor. The MFR was 16.9, which is 300 %
higher than MFR in Comparative Example 2a, where donor D was used. The details and
results are shown in Table 2.

Comparative Example 3a

This example was carried out in accordance with Comparative Example 1a, with the
exception that the hydrogen amount was 1500 mmol. The MFR was 210. The details and
results are shown in Table 2.

Comparative Example 3b

This example was carried out in accordance with Comparative Example 3a, with the
exception that donor C was used as external donor. The MFR was 470, which is 220 %
higher than MFR in Comparative Example 3a, where donor D was used. The details and
results are shown in Table 2.

Table 2. The Results of the Comparative Examples

Table 2		Comp. Ex 1a	Comp. Ex 1b	Comp. Ex 2a	Comp Ex 2b	Comp. Ex 3a	Comp. Ex 3b
Time	min	60	60	30	30	60	60
Hydrogen	mmol	13	13	200	200	1500	1500
Donor	type	D	C	D	C	D	C
Productivity	kg PP/ g cat	21	18,8	23,8	18,7	38,9	29,2
MFR	g/10 min	0,36	0,70	5,3	16,9	210	470
XS	w-%	1,2	1,8	1,1	1,6	1,7	2,7
FTIR Isotacticity	%	96,9	94,6	100,4	98,8	104	103,7
Tm	°C	168,5	165,1	165,8	163,3	161,3	160,2
Flexural modulus	Mpa	2050	1710		1600	1720	1770

Discussion of results

The difference between the catalyst behaviour used in the examples of the invention and

the typical, supported Ziegler Natta catalyst used in the comparative examples is best seen
in Figures 1 and 2. The catalyst used in the examples of the invention show no difference
in hydrogen response independent on whether the external donor is donor C or donor D,
Figure 1. For the typical, supported Ziegler Natta catalyst used in the comparative

5   examples, donor C gives 100-300 % higher MFR than donor D at a certain hydrogen
concentration, Figure 2.

From Table 1 it is also evident that even if the catalyst in this invention shows the same
hydrogen response for donor D and donor C (MFR values are essentially at the same

10   level), the normal desired differences in polymer properties caused by using different
donors, D and/or C, are maintained as was the target. Donor D gives clearly higher
isotacticity, lower XS, higher T_m and higher stiffness than donor C. The normal differences
caused by using donor D and donor C can be seen also with the typical, supported Ziegler
Natta catalyst used in the comparative experiments and summarized in Table 2. However,

15   with the comparative catalysts, as can seen in Table 2, there are big variations between
donor C and D in hydrogen response, which is shown by the huge differences between the
MFR results, when the hydrogen feed is kept constant.